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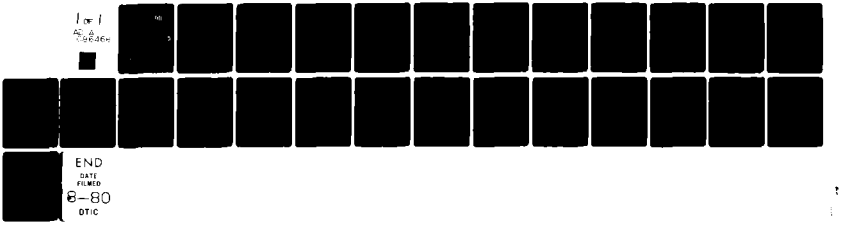

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Department of Applied Physics.

U.S. Army Research and Development Group (Europe)

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INTEGRATED THIN FILM DC SQUIDS

Principal Investigator: Dr. G.B. Donaldson

Grant number: 76 - G - 079

Final Report - to 31 October, 1979.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A new thin film low temperature facility has been developed and used on three aspects of DC SQUIDS. 1) Ellipsometric studies of RF sputter - etch oxidation of Pb-In alloys and Nb. 2) Nb-NbO_x-Pb/In Josephson junctions as planar SQUIDs 3) Nb-NbO_x/Au-Nb junctions as SQUIDs, inferior noise performance.

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INTEGRATED THIN FILM DC SQUIDS

Final Report

Abstract

A new thin film low temperature facility has been developed and used for work on three aspects of DC SQUIDs. First, ellipsometric studies have been made of the process of RF sputter-etch oxidation of Pb-In alloys and of Nb. Second, structures based on Nb-NbO_x-Pb/In Josephson junctions have been produced and successfully operated as planar SQUIDs with a flux noise of $8.5 \times 10^{-5} \Phi_o\text{-Hz}^{-\frac{1}{2}}$. Finally, all Nb structures, based on Nb-NbO_x/Au-Nb junctions, have been operated as SQUIDs though so far with inferior noise performance ($5 \times 10^{-2}\ \Phi_o\text{-Hz}^{-\frac{1}{2}}$). Work is proceeding on the production of integrated magnetometric structures using photolithographic techniques to etch planar flux-transformers and the associated components.

Contents

INTEGRATED THIN FILM DC SQUIDs

Final Report

1. General

The grant which is the subject of this report ($12,600 for the period October 1976 - October 1979) was awarded to provide support for a graduate student who would work with the DC SQUID (Superconducting Quantum Interference Device) group at Strathclyde University. The principal funding for the SQUID programme (totalling about £60K) has been provided by Strathclyde University and by the British Science Research Council, on grants which end on 31 May 1980, and for which final reports are due in September 1980.

The student appointed under this grant (Miss F.M. Livingston) has completed her experimental work. However her Ph.D. thesis is not expected until May 1980, and no papers based on it can be prepared until she has submitted it. Meanwhile, research on the programme is continuing, with the further U.S. Army E.R.O. support of $5,000 for the period to October 1980 (DAJA-37-79-M-0314). It will be seen that the report (covering the period to last October) can be 'final' only in a rather legalistic sense. A full review, especially of Miss Livingston's conclusions, will be made in the final report on the new contract. In this document, we therefore make a general report on the progress of the SQUID group up to 31 October 1979.

2. Development of Superconducting Device Facilities

Work began in May 1976 when I and Dr. Stern took possession of a newly emptied suite of rooms (area $80m^2$). The detailed development of the personnel of the group is given in Appendix B: we here mention only that at October 1979, the group contained two faculty, one research fellow, three graduate students and one technician, of whom four were associated with the SQUID work. We also wish to record, with great sadness, the loss, by his untimely death in 1977, of Dr. Stern's involvement: this seriously set back the research program, probably by at least a year.

By October 1979, the group was equipped with a range of thin film equipment, including conventional evaporators, DC sputtergun, a clean high vacuum (5×10^{-8} torr) evaporator, and a low power RF sputter etching facility. It had limited photolithographic capabilities, and was making some progress towards achieving modest 'clean room' capability. On the cryogenic side, a reasonable range of cryostats, both fixed and portable, had been acquired, together with pumps and other standard equipment, making it possible to cover the temperature range from 1.2 K to 300 K. One evaporator was equipped with ellipsometry facilities capable of measuring optical parameters of metal surfaces and of thin oxide films grown on them either by thermal or by RF oxidation: from these parameters oxide thickness can be obtained with a resolution of about 0.2nm in a typical thickness of 1-10nm.

A DEC-LAB 11-03 minicomputer with 32 K of memory had also been obtained and programmed for use as a signal averager and spectrum analyser of SQUID signals.

3. Research to October 1979

Thin Film DC SQUIDs are extremely sensitive detectors of magnetic field, and are based (see Fig. 1) on a loop of superconductor containing two Josephson junctions. These junctions are formed from two thin film electrodes separated by an oxide layer between 2 and 5 nm thick. With appropriate electronics, these devices can resolve fields down to 10^{-14} Tesla and below. Applications have begun to appear in many branches of science and engineering, examples being geophysics, human physiology, submarine communications and physical chemistry. Very useful broad reviews of the field are available [1], and we will not attempt to summarise them here. Our efforts in DC SQUIDs have had three thrusts (a) an attempt to understand and control the growth of oxides suitable for tunnel barrier formation on Pb-In and on Nb films (b) a program to develop the capability of making Clarke [2] type planar SQUIDs and to design them into magnetic gradiometers and (c) an attempt to produce an all-niobium thin film SQUID. With each of these aspects we had some success by October 1979, though, as explained in §1, we expect further progress by October 1980. We here report separately on each of these sub-programmes, but it is worth reiterating that the ERO support was essentially wholly applied to the graduate student associated with part A below.

A. R.F. sputter-etch oxidation and ellipsometric monitoring of oxide growth.

The sputter-etch procedure for Josephson barrier formation was first described by Greiner [2]. It uses a 13.6 MHz RF discharge run in a low pressure of oxygen (5 - 50 millitorr) to produce rapid oxidation of a metal film surface. The growth rate decreases exponentially as the oxide thickens, and ultimately a dynamic equilibrium is reached between oxide growth and the sputtering away of oxide by activated gas ions. Using

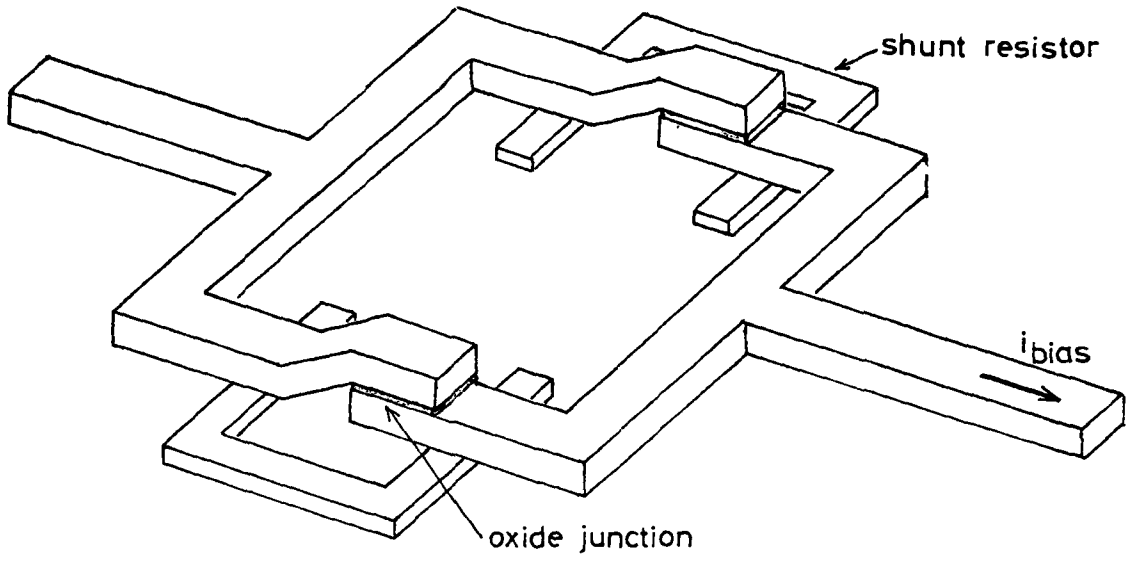


Fig. 1(a): Schematic of a tunnel junction D.C. SQUID structure. Two oxide junctions are connected in parallel to form a superconducting loop whose area is typically $1mm^2$ and whose inductance is about 1nH. The junction areas are of the order of 10-100μm square and there are usually shunt resistances of order 0.5Ω across them.

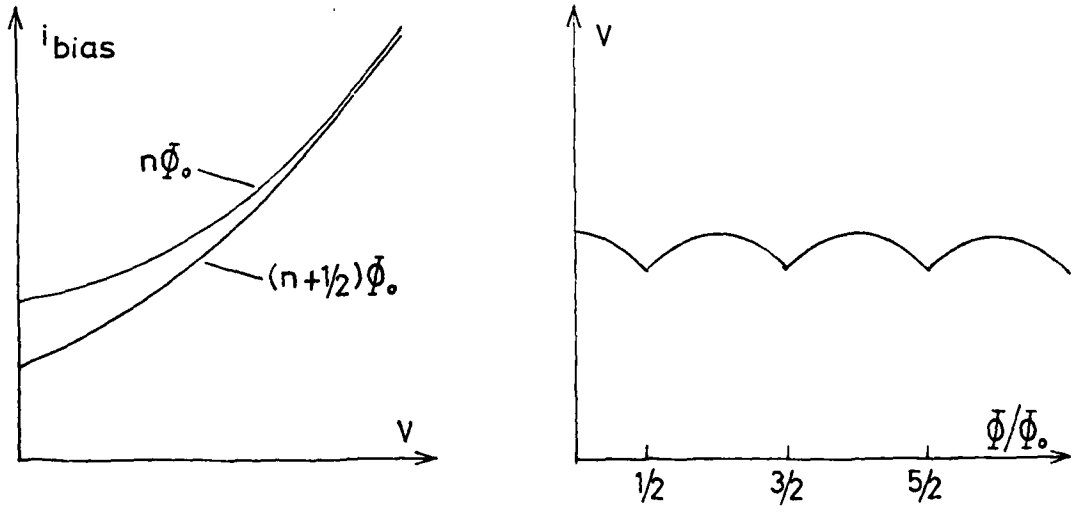


Fig. 1(b): Current-voltage characteristics under various applied magnetic fields. When i_{bias} exceeds the critical current, the voltage V is a periodic function of the flux Φ applied to the loop. The period is Φ_o (2×10^{-15} Wb) which corresponds to 2×10^{-9} Tesla for a loop area of 1mm x 1mm.

this technique, Greiner and his colleagues produced very successful Josephson junctions, intended for computer applications, on Pb-In alloy films.

We have explored the Greiner method with a view to producing barriers on evaporated Pb-In films and on sputtered Nb films suitable for SQUID junctions. We used apparatus based on a commercial evaporator, in which we included a home-made low power 13.5 MHz sputter gun for sputter etching and a simple ellipsometer based on a helium-neon laser with polariser and analyser of 0.01^{o} resolution. The standard procedure of ellipsometry yielded the refractive index components ($\bar{n}$ and $\bar{k}$) for unoxidised substrate films, and the refractive index (n) and thickness (d) of transparent oxide films formed on them. The precision of thickness measurements is about 0.2 nm in a typical total of 1-10 nm.

We have studied the oxidation of Pb-In alloys across the whole range of alloy concentration. The results show monotonic oxide growth for most films, with saturation thicknesses (3.0 - 9.0 nm) being reached within ~ 10 min for small In concentrations, and ~ 40 min for higher In concentrations. For films of 26-36 atomic % nominal In concentration, the oxide thickness rose to an intermediate maximum before decreasing to the equilibrium value. The In_2O_3 content of the oxide layer was disproportionately large and, in particular, was 100% for all alloys containing more than 28 atomic % of indium.

A brief account of these results was given in a paper at the 1978 conference on superconductive electronics [3], (copy attached to this report). Analysis of the data has continued, and by October 1979, it was shown that the 'intermediate maximum' behaviour could plausibly be modelled by a discrete columnar model of separate PbO and In_2O_3 phases, with growth of a particular species (say PbO at the Pb-In surface) taking place as a result of parallel diffusion of oxygen atoms down both PbO and In_2O_3 columns. It is expected that this work will be completed in 1980.

The work on sputtered Nb films has had less satisfactory results so far. It has certainly proved possible to form NbO_x barriers by RF oxidation, and as with Pb-In, to make satisfactory Josephson junctions using them. However, the results have not been reproducible, and we have found that no advantage of predictability is obtained over the cruder thermal techniques described later (see §C). The problems appear to be in the high chemical affinity of niobium for oxygen, water vapour and even nitrogen, and with the lack of sophistication of our equipment. The Nb films have to be sputter-deposited in a different chamber from the one in which RF oxidation is carried out, and it is clear from the variability of superconducting behaviour (see §B) that the nature of these films is critically dependent on the levels of minute quantities of N_2 and H_2O in the sputtering gas (Argon), and also on the sputtering rate. All of these are difficult to control. As a result, the fundamental chemical properties of the Nb film transferred to the oxidation chamber is unpredictable. Moreover, the transfer exposes it to the air, and causes it to acquire a fairly thick uncontrolled layer of oxide.

The RF oxidation and ellipsometry procedures are thus begun on a film with uncertain oxidation properties, which is covered with an initial 'oxide' layer of doubtful constitution. With Pb-In such an initial oxide layer presents little difficulty, because RF sputtering in Argon produces cleaning without oxidation and strips the sample back to the bare alloy. With niobium, however, the outgassing of the evaporator and the liberation of water vapour by the operation of the gun produced sufficient reactive gas to negate the stripping action of the argon etch and it was usually impossible to obtain a bare metal surface. The values of $\bar{n}$ and $\bar{k}$ could therefore not be obtained precisely, so that calculations of precise oxide thickness (d) and index (n) could not be made during subsequent barrier formation. The properties of our barriers were therefore unpredictable.

By substantially increasing the RF power used in the sputter-etch we should be able to completely clean our Nb films before oxidising them. This could be expensive however, and has not yet been tried.

Fuller details of the niobium oxidation work will be contained in Miss Livingston's thesis.

B. Nb-NbO_x-Pb/In DC SQUIDs

By October 1979 we had achieved the ability to produce Clarke type [3] structures in a planar geometry and to operate them at a comparable (indeed slightly better) noise level than those previously reported. We discuss our techniques in this section. None of the work was published by 1979, but some will be during 1980.

(1) DC sputtering of superconductive niobium

Niobium is an extremely desirable material for thin film superconducting device material because it is tough, undamaged by the inevitable water condensation that is associated with thermal cycling between cryogenic and room temperature environments, and because its critical temperature (normally 9.1K) is well suited to the usual cryogenic temperature regime (4.2K). It is very refractory, though, (MP 2420^{o}C), and high power electron beam techniques are needed to evaporate it at a reasonable rate. These in turn predicate 10^{-8}torr vacuum facilities, and high pumping speeds to cope with outgassing during electron-gun operations. This is expensive and time consuming, and makes the use of a sputtering technique for film deposition highly desirable. We have used a SLOAN S310 DC sputtergun, mounted on a conventional 10^{-5} torr vacuum system. Because the sputtering is performed in a gas (Argon) rather than in vacuum, there are high risks that the sputtered material will contain impurities. These tend strongly to reduce the T_c of niobium, and can easily render it non-superconducting. We find the following conditions to be necessary to guarantee superconductivity in the sputtered Nb films:

(a) System initially pumped out to $\leq 2 \times 10^{-6}$ torr, with LN_2 trap to remove water vapour.

(b) High purity Argon - we use 'zero-grade' gas with specified impurity levels for the 'risk' gases, viz O_2, H_2O<1ppm, N_2<8ppm.

(c) Deposition rate $\geq$ 75 nm-min^{-1} (typical conditions are - Argon pressure 10m-torr, sputter gun voltage and current 345V and 3 amp, and sputtering time 3½-5 minutes.

(d) Sputter gun and substrate not more than 5-7 cm apart. No materials in path of sputtered Nb that might outgas or decompose under bombardment: plastics and adhesives must be avoided.

(e) Pre-sputtering is necessary (typically 2 minutes at 3 amps) with shutter between gun and substrate closed. Shutter must be opened, deposition completed and shutter closed again, before the gun is turned off.

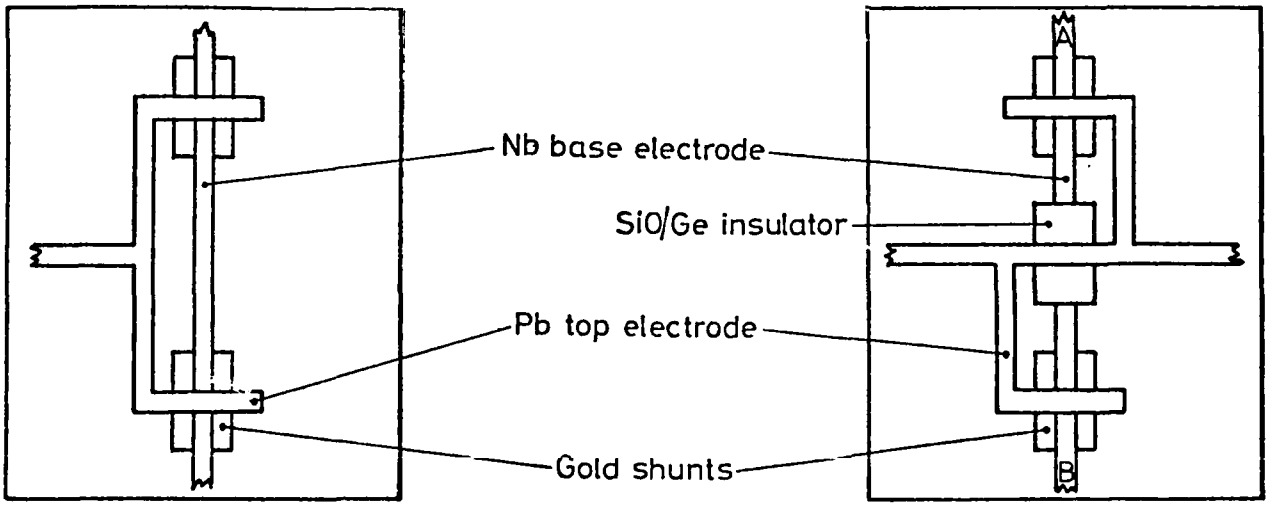
(f) The substrate (soda or Corning 7059 glass) need not be heated during deposition, though in our system its proximity to the gun ensures that it gets very warm. It must be scrupulously clean, however, to ensure good adhesion. We (i) ultrasonically clean in concentrated detergent (DECON 90) heated to boiling point (ii) rinse in deionised water, drain and transfer to boiling ultrasonically agitated acetone bath and (iii) flame to dull red heat before cooling in air.

Good niobium films have very high adhesion to their substrates. They will only go superconducting if their 300K: 77K: 10K conductance ratios are better than 1: 1.25: 2.5. "Bad" specimens have room temperature resistances up to 40 x those of "good" specimens, and may actually increase their resistance on cooling.

SQUID construction

SQUIDs with Nb-NbO_x-Pb junctions are made by deposition through out-of-contact masks. The masks are etched in stainless steel foil 25μm thick; the required patterns are drawn 20 x true size, photo-reduced and transferred to the stainless foil using photolithography with spun-on Kodak KPR3 photoresist. The foil is then etched, mounted on a backing support and polished flat.

SQUIDs have been made in both the magnetometer configuration of Fig. 2 and the gradiometer design of Fig. 3. The latter has no uniform field response and is used to sense current flow in the track AB. Thin-film deposition starts with gold shunts some 10nm thick (with a 5nm chrome underlay to improve adhesion). Next, a Niobium base electrode 200nm thick is deposited using the sputtergun. The junction oxide is next formed by oxidation in air at 130-150°C for 10-15 minutes. For the gradiometric SQUIDs (Fig. 2), the insulation for the central crossover is deposited after oxidation (100nm SiO plus 500nm Ge). Care is needed here to prevent these insulator materials contaminating the junction areas. The final evaporation forms the top electrode (Pb + 5% In, 300nm thick). Indium contact pads are then cold-welded into place and a coat of KPR3 is spun on to protect the finished device. Without this protection, moisture (deposited primarily as hoar frost) tends to react with the lead and dissolve away the top electrode.



Magnetometer style SQUID

Fig. 2

Gradiometer style SQUID

Fig. 3

The SQUIDs are used with 100 KHz modulation in the circuitry depicted in Fig. 4. A key feature of this is the cooled LC matching circuit ($Q \approx 100$) that matches the low impedance of the SQUID to the high optimum input impedance of the following amplifier. The SQUID is DC biased just above its critical current, where the periodic modulation (by applied flux) of the voltage across the SQUID is phase sensitively detected. With switch S closed, the system operates in a flux-locked loop (FLL) mode by locking onto a minimum in the 100 KHz modulation.

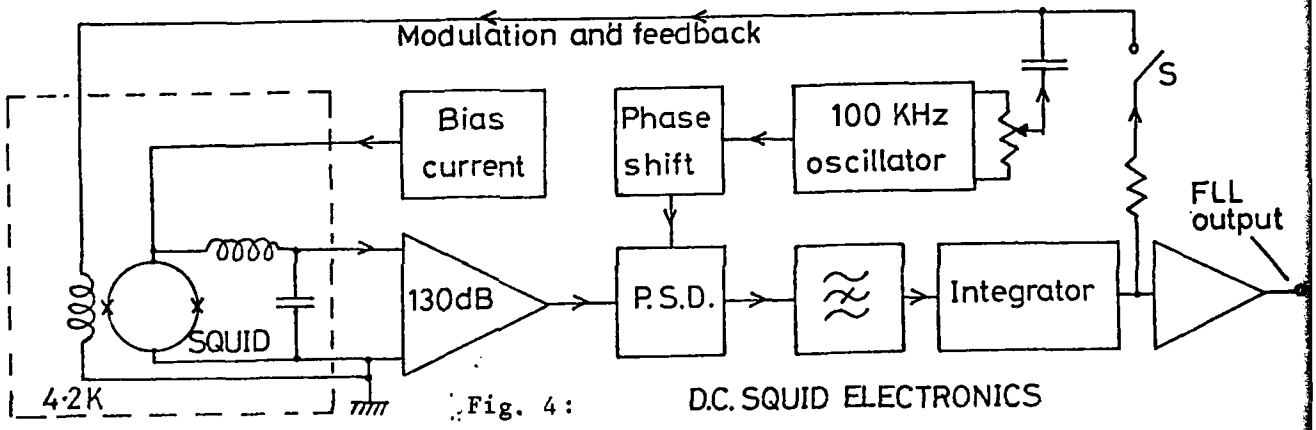


Fig. 4: D.C. SQUID ELECTRONICS

Operation

Summarised below are typical system parameters (as at October 1979) for a magnetometer style of SQUID 2mm long by 0.27mm wide formed from tracks 100μm wide.

Normal state resistance 0.34 ohm

Critical current 0.6μA, optimum bias current 1.6μA

Mutual inductance between track AB and SQUID 0.91nH

Self inductance of SQUID 2.0 ± 0.2nH

FLL output 0.23 V Φ_o^{-1}

FLL output noise level ≤ 200μV peak-to-peak in bandwidth 0.1 to 12Hz, which corresponds to an equivalent flux noise $S_\Phi^{\frac{1}{2}}$ of 8.5×10^{-5} $_o Hz^{-\frac{1}{2}}$.

Work is now proceeding on identifying the prime sources of noise in these SQUIDs (which are the best planar SQUIDs reported) and to reduce them. We have also begun to integrate them into larger gradiometer structures, and are devoting effort to studying how to optimise such structures.

C. All niobium structures

We have directed considerable effort towards producing tunnel junctions and d.c. SQUIDs using niobium rather than lead-indium as a counter electrode for the following reasons:-

(a) Niobium films are extremely resistant to mechanical damage i.e. abrasion on ice-crystal formation, and require no further protection.

(b) Niobium films suffer no adverse reactions with most chemicals, in particular atmospheric water vapour.

(c) Solubility of oxygen is much lower in niobium than in lead; this should lead to greater stability at the niobium oxide/counter-electrode interface.

(d) The excellent adhesion of niobium to glass should remove any problems of hillocking of the films leading to alteration of junction characteristics with thermal cycling.

(a) Use of Niobium Oxide/Gold as a Tunnel Barrier

Direct sputtering of niobium onto oxidised niobium leads almost inevitably to superconducting shouts across the tunnel junction. However, developing an idea of Hawkins,[5] we find this problem may be circumvented by deposition of very thin layers of copper, silver or gold on top of the oxide prior to deposition of the counter-electrode. Thicknesses of metal required to ensure a high yield of non-shorted junctions are found to be:-

(a) Copper: $\geq 10\text{Å}$

(b) Silver: $\geq 6\text{Å}$

(c) Gold: $\geq 3\text{Å}$

We believe the normal metal acts 'protectively' by filling defects in the oxide and preventing reduction of the oxide by incoming niobium. It is noteworthy that by comparison with copper and silver, a much sparser layer of gold (high atomic weight) is required for success. Also, of the 3 metal layers, copper and silver tend to impair adhesion of the Nb counter-electrode. Gold alone does not, due, we believe, to its deposition as a particulate formation. This, coupled with the very small quantity required (which we believe should lead to lower noise from these junctions) makes it the obvious choice as a protective metal.

(b) Fabrication

Large area (440 μm x 440 μm) tunnel junctions (used to determine junction characteristics at current densities similar to those used in the SQUIDs) and d.c. SQUID magnetometers are fabricated as follows.

(i) Corning 7059 glass slides are degreased ultrasonically in carbon tetrachloride for 15-20 minutes, then baked at 190°C for 20 minutes.

(ii) SQUID shunt resistors are laid down through a machined aluminium mask by evaporating 15Å chromium and 30Å gold at a pressure of 10^{-6}T.

(iii) Base niobium electrodes are sputtered through an aluminium mask.

(iv) Although thermal oxidation (§B) can be used with this procedure, it is not usable in conjunction with photo-lithographic production of the base electrode as described below. An alternative plasma discharge scheme has therefore been adopted, in which the evaporator vessel is first evacuated to $<10^{-6}$ torr to reduce the effects of outgassed water vapour. Dehydrated air is admitted to 100 millitorr, and a

D.C. plasma discharge run for 2-8 minutes. The specimen is mounted near the centre of the chamber on a glass support.

(v) 3.5Å of gold is then deposited at 10^{-6} torr, at a rate not lower than 0.5Å per second.

(vi) The Nb counter-electrode is sputtered under identical conditions to the base electrode.

(c) Junction and SQUID Properties

Using the above procedures, we have produced series of junctions and D.C. SQUIDs. In common with other form of niobium/niobium junction, we note quasi-particle currents in excess of those predicted by the Ambegaokar-Baratoff[6] relations; however, this is not large enough to affect the performance of the SQUIDs. Junction yield is very high (> 75%), and, apart from an initial rise in critical current after the first thermal cycle to 4.2K (due, we believe, to stabilisation of the oxide barrier), critical current stability with thermal cycling is excellent. Storage over several months at room temperature leads to an increase in critical current. Due to the difficulties concerning barrier thickness control mentioned above (and now overcome), we have not so far optimised the critical currents of our SQUIDs; thus those SQUIDs produced so far have had considerably poorer noise properties than our SQUIDs using lead-indium counter-electrodes. Typically, our devices have had critical currents of 30μA and inductance of 0.8nH, and have effectively been self-shunted with a resistance of 50mΩ. The measured noise has typically been 5×10^{-2} Φ_o-$Hz^{-\frac{1}{2}}$: however this is in agreement with the predictions of SQUID noise theory, and should be greatly improved once our fabrication procedures give us more control over the various junction parameters.

Further work, to optimise the barrier thickness, is in progress.

(d) Photo-lithographic techniques

Given the availability of all-niobium thin film SQUIDs, the fabrication of complex thin film niobium structures as magnetic pick-up coils into which the SQUIDs can be incorporated is highly desirable. It follows that methods based on photolithography, using circuit patterns projected on to photo- together with suitable solvents and etches, are very desirable. This presents new difficulties, however, because procedures for dealing with niobium are not well established, and techniques capable of coping with all of the metals we employ have not been described at all. For example - a known niobium etchant is an appropriate mixtures of $HF/HCl/HNO_3$ in water, but this would strongly attack any chromium which had been previously deposited. New procedures and sequences must therefore be explored. We have had some success in this, and will describe our detailed procedures in a later report.

Conclusions

We have made considerable progress in establishing the conditions necessary for SQUID preparation, and have had substantial success in producing low noise devices. Very rugged all niobium structures have been developed, though further work is necessary to improve their noise performance. In the coming year we expect to produce a number of integrated structures with a view to use in a number of applications such as magnetic anomaly detection.

References

1. J. Clarke, SQUIDs for Low Frequency Applications, Chap. 3, pp. 67- in 'Superconductor Applications': SQUIDs and Machines, B.B. Schwartz and S. Foner, Eds., Plenum Press, New York, 1977.

M.B. Ketchen, J. Clarke and W.M. Goubau, in 'Future Trends in Superconductive Electronis' (B.S. Deaver, C.M. Falco, J.H. Harris and S.A. Wolf, Eds.), A.I.P. Conf. Proc 44, 32, (1978).

2. J.H. Greiner, J. Appl. Phys. 42, 5151, (1971).

J.H. Greiner, J. Appl. Phys. 45, 32, (1974).

3. G.B. Donaldson and H. Faghihi-Nejad, in Future Trends in Superconductive Electronis (B.S. Deaver, C.M. Falco, J.H. Harris and S.A. Wolf, Eds.), A.I.P. Conf. Proc. 44, 407 (1978). (Copy attached to this report.)

4. J. Clarke, W.M. Goubau and M.B. Ketchen, J. Low Temp. Phys. 25, 99 (1976).

5. G.A. Hawkins and J. Clarke, J. Appl. Phys. 47 1616, (1976).

6. V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. 10, 486 (1963).

Appendix A

Financial report

With the exception of a small sum used by the grantholder for U.K. travel, the entire sum ($12,600) was devoted to the support of Miss Livingston. She was remunerated precisely according to the rules for Science Research Council research studentships. The incidence of costs was as follows:

	1976/7 £	1977/8 £	1978/9 £
Student's Salary	1274	1455	1610
Daily Travelling Expenses	78	137	136
University Fees	333	795	1132
Other travel	16	12	
	1701	2399	2878

Appendix B

Low Temperature Group - Personnel Associated with SQUID work

	Joined	Left	Proportion of time on this work
Dr. G.B. Donaldson (Lect./Sen. Lect.)	1976	-	30%
Dr. F. Stern (Lecturer)	1976	1977 (deceased)	30%
Dr. C.M. Pegrum (Research Assistant Teaching Fellow (1977/78))	1976	-	80%
Miss F. Livingston (Research Student)	1976	1979	100%
Mr. R. Bain (Research Student)	1977	-	100%
Dr. N.A. Lockerbie (Lecturer)	1979	-	10%

Appendix B

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Dr. G.B. Donaldson (Lect./Sen. Lect.)	1976	-	30%
Dr. F. Stern (Lecturer)	1976	1977 (deceased)	30%
Dr. C.M. Pegrum (Research Assistant Teaching Fellow (1977/78))	1976	-	80%
Miss F. Livingston (Research Student)	1976	1979	100%
Mr. R. Bain (Research Student)	1977	-	100%
Dr. N.A. Lockerbie (Lecturer)	1979	-	10%

Future Trends in Superconductive Electronics (B.S. Deaver, C.M. Falco, J.H. Harris and S.A. Wolf, Eds.), A.I.P. Conf. Proc. 44, (1978).

RF SPUTTER ANODISATION OF LEAD-INDIUM FILMS STUDIED BY ELLIPSOMETRY

G.B. Donaldson, Department of Applied Physics, University of Strathclyde, Glasgow, Scotland.

H. Faghihi-Nejad*, Department of Physics, University of Lancaster, England.

ABSTRACT

We have used simple ellipsometric equipment to study the RF sputter oxidation of a series of lead-indium alloy films containing from 0-46 atomic % In. Our results show monotonic oxide growth for most films, with saturation thicknesses (3.0 - 9.0 nm) being reached within ~ 10 min for small In concentrations, and ~ 40 min for higher In concentrations. For films of 26-36 atomic % nominal In concentration, the oxide thickness rose to an intermediate maximum before decreasing to the equilibrium value. The In_2O_3 content of the oxide layer was disproportionately large and, in particular, was 100% for all alloys containing more than 28 atomic % of indium.

INTRODUCTION

Lead-indium alloy films have shown considerable promise as materials on which to grow Josephson-junction oxide barriers. Eldridge et al[1] have used ellipsometry to study the developments of oxide layers grown thermally in atmospheres of oxygen over periods of up to five hours. We have looked ellipsometrically at similar films on which the barrier has been grown using the technique described by Greiner[2], in which a low power RF discharge in oxygen is used to produce a dynamic equilibrium between the rate at which oxide is removed by sputtering and that at which new oxide is being formed by activated oxygen atoms. The work was undertaken to further elucidate a technique of ours[3] which is known to produce junctions with excellent critical current characteristics.

APPARATUS AND METHOD

We used Joule-heated boats in a standard evaporator (Fig.1) to deposit films on Corning 7059 glass substrates which were mounted on a RF target suspended from the bell jar top plate.

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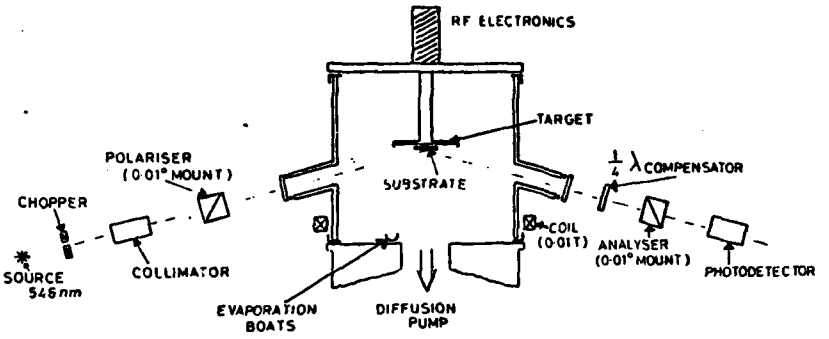
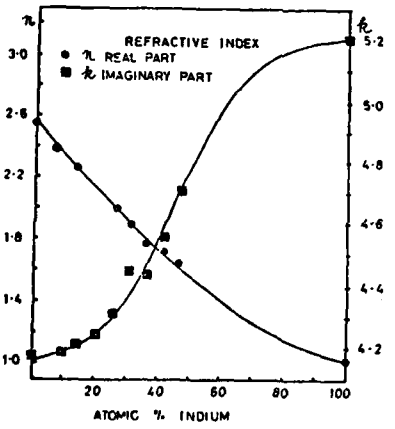


APPARATUS LAYOUT

Figure 1

Polarized green (546 nm) light entered the vacuum chamber through a strain-free window mounted on a side arm, was incident on the substrate at 70°. The reflected beam passed through another strain-free window on a second side arm, and was detected with a photomultiplier after passing through a quarter-wave compensator and an analyser. Extinction settings of the polariser and analyser could be made to 0.04°, and from them we determined Δ and ψ, the parameters describing the change which reflection produces in the ellipticity of the polarisation of the incident beam. For bare metal surfaces we used Δ and ψ in standard formulae[4] to determine the real (n) and imaginary (k) parts of the refractive index; in the case of oxide layers we determined their refractive index (n) and and thickness (d) by assuming that their absorption was zero and applying the approximate formulae of Archer[5].



MEASURED OPTICAL CONSTANTS OF LEAD–INDIUM ALLOYS

Figure 2

Our films were evaporated at 5×10^{-6} torr as described by Emmanuel et al.,[3] with indium being deposited first and lead overlaying it. The total thickness was about 200 nm and the relative thicknesses of the two layers represented the required alloy ratio. We made ellipsometric measurements of n and k on the films immediately after evaporation and for pure lead and pure indium found good agreement with other workers[1,2]. On alloys, however, we found that Δ and ψ varied in a manner consistent with the diffusion of

indium into the lead layer, reaching equilibrium after about 30-90 minutes at room temperature. The limiting n and k values fit a roughly linear interpolation, based on nominal concentration, between lead and indium. (Fig.2) This suggests that the diffusion process homogenises the film completely.

To prepare insulating layers, we admitted 10-100 millitorr of pure oxygen. After 5 minutes, during which a few angstroms of oxide were observed to form, we started an RF discharge between the target (as anode) and ground, using a 10^{-2}T axial magnetic field to concentrate the discharge. A simple single valve 13.5 MHz oscillator circuit was used, and the power incident on the films was of order 0.6-4 mW-cm^{-2}: the substrate temperature rise was negligible. The discharge was stopped occasionally to make ellipsometric measurements of n and d. No oxide growth could be detected during these intermediate intervals.

OXIDATION RESULTS AND DISCUSSION

Thickness of oxide layer

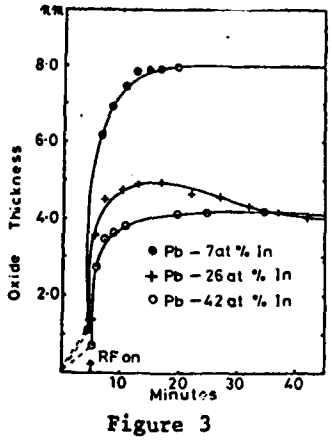


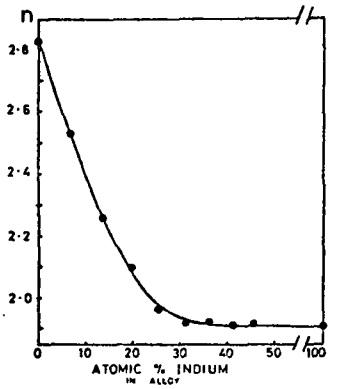
Figure 3

Figure 3 shows the growth of oxide for three films of different indium concentrations under typical pressure and power conditions. The top curve is characteristic of films containing 0-20 atomic % of indium, with initial growth rates of about 3.5 nm-min^{-1}, decreasing monotonically until a saturation thickness X_s is reached after about 15 minutes. Simple behaviour is also observed with concentrations of indium greater than 35%, and with pure indium films: initial growth rates are slower and the time constants are longer than in the case of lead-like films, with 40 minutes being required to reach saturation in the case of pure In. The most striking behaviour, however, is observed in the range 26-36 atomic % In (middle curve). Here, for power levels greater than 2 mW-cm^{-2}, the oxide thickness rises with a short time-constant and reaches an intermediate maximum before decreasing to its equilibrium value. We do not fully understand the mechanism for this behaviour, but believe that it may be due to preferential sputtering of islands of lead oxide such as are postulated by Eldridge et al.[1] in their thermal oxidation work. After initial rapid removal of material from lead-rich islands the controlling time constant could become the rate at which lead can diffuse through the lead-indium alloy or indium oxide. It is not clear, however, how this can account for a decrease in the measured

barrier thickness.

The saturation thickness X_s was found to increase with increasing oxygen pressure: for example in a specimen of 8 atomic % In subjected to 5 mW-cm^{-2} power, the first thickness was 74 nm in a 10-millitorr discharge, but this increased to 84 nm when the pressure was raised to 50 millitorr. When the pressure was returned to 10 millitorr, X decreased again over about 20 minutes to the 74 nm value, demonstrating the true dynamic equilibrium of the sputter-etch procedure. The dependence of X_s on RF power was not clear-cut however: in general for lead-rich alloys as in Fig.3 (top), X_s was decreased by increasing the power, but for alloys with more than 30 atomic % In. X_s was a weakly increasing function of RF power under our operating conditions.

Oxide composition



REFRACTIVE INDEX OF OXIDE FILM

Figure 4

The refractive index (n) of the anodised layer was found to vary very little during oxidation, and in Fig. 4 we show how it varied with indium concentration. The data agree well with those of Eldridge et al.[1], obtained in their natural oxidation work at 760 torr. In particular the oxide index decreases from that of pure PbO at a zero indium concentration to that of pure In_2O_3 at all indium concentrations above 30%. This conclusion - that the effective composition of the barrier is much enriched In_2O_3 - is in agreement with evidence from ion scattering spectroscopy [3] and Auger spectroscopy. [6]

CONCLUSION

RF sputter-etching is an effective and rapid way of producing oxide barriers of controlled thickness on lead-indium alloy films. Only inexpensive equipment is needed. The composition of the oxide films is similar to that obtained when the growth takes place more slowly by natural oxidation. The mechanism of oxidation is not simple, however, as is shown by the intermediate maximum which is seen as oxide grows on films of intermediate composition (26-36 atomic % In), and which possibly arises by preferential sputtering from small islands of enriched Pb composition with consequent inter-grain Pb diffusion within the film.

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REFERENCES

1. J.M. Eldridge, D.W. Dong and K.L. Komarek, J. Electron. Mater. 4, 1191 (1975).

2. J.H. Greiner, J. Appl. Phys. 45, 32, (1974) and 42, 5151 (1971).

3. A. Emmanuel, G.B. Donaldson, W.T. Band and D. Dew-Hughes, I.E.E.E. Trans. Magnetics, MAG-11, 763 (1975).

4. See for example M. Born and E. Wolf, Principles of Optics, 2nd ed., Pergamon, London (1964).

5. R.J. Archer, J. Electrochemical Soc. 104, 619 (1957).

6. N.J. Chou, S.K. Lahiri, R. Hammer and K.L. Komarek, J. Chem. Phys. 63, 2758 (1975).